

Tuning of Exciton States in a Magnetic Quantum Ring

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We have studied the exciton states in a CdTe quantum ring in an external magnetic field containing a single magnetic impurity. We have used the multiband approximation which includes the heavy hole - light hole coupling effects. The electron-hole spin interactions and the $s, p - d$ interactions between the electron, hole and the magnetic impurity are also included. The exciton energy levels and optical transitions are evaluated using the exact diagonalization scheme. We show that due to the spin interactions it is possible to change the bright exciton state into the dark state and vice versa with the help of a magnetic field. We propose a new route to experimentally estimate the $s, p - d$ spin interaction constants.

Electronic properties of planar nanoscale semiconductor structures, such as quantum rings (QRs) [1] and quantum dots (QDs) [2] have enjoyed widespread attention in the past few decades due to their novel fundamental effects and for potential technological applications. Experimental advances in creating these structures from a two-dimensional electron gas by using suitable confinements have resulted in confirmation of several theoretical predictions in these systems [3, 4]. It has been realized lately that QD doped with a single magnetic impurity [5–7] has great potential to contribute significantly in the burgeoning field of single spin manipulation [8], which will eventually lead to important contributions in quantum information processing. Quite naturally, quantum dots, in particular the CdTe QDs containing a single Mn atom has been widely studied in the literature [9]. It has been proposed that magnetic doping of QDs provides an interesting route to magnetism in the QDs that can be tuned [10]. Against the backdrop of these important developments, no such studies involving a quantum ring have been reported yet in the literature. Recently, CdTe QRs have been realized experimentally [11]. Here we report on our studies of the exciton states in a CdTe QR in a magnetic field, containing a single magnetic impurity. We have found that, due to the resulting spin interactions the bright exciton state can be changed to the dark state and vice versa, with the help of an applied magnetic field. Additionally, we propose here an experimental means to estimate the $s, p - d$ spin interaction constants.

We study the exciton states in a CdTe quantum ring containing a single manganese magnetic impurity (Mn) and subjected to a perpendicular magnetic field. Usually, the thickness of the ring is smaller than the radial dimensions. Therefore, our system can be considered as quasi two-dimensional, with internal radius R_1 and the external radius R_2 . The electron and hole are always in the ground state for the z direction. We chose the confinement potential of the quantum ring in the radial direction with infinitely high borders: $V_{\text{conf}}(\rho) = 0$ if $R_1 \leq \rho \leq R_2$ and infinity outside of the QR. The Hamiltonian of the

system can then be written as

$$\mathcal{H} = \mathcal{H}_e + \mathcal{H}_h + V_{\text{eh}} + \mathcal{H}_{\text{eh}} + \mathcal{H}_{\text{s-d}} + \mathcal{H}_{\text{p-d}} + \mathcal{H}_{\text{Mn}}, \quad (1)$$

where $\mathcal{H}_{\text{s-d}} = -J_e \delta(\mathbf{r}_e - \mathbf{r}_{\text{Mn}}) \boldsymbol{\sigma} \mathbf{S}$ and $\mathcal{H}_{\text{p-d}} = -J_h \delta(\mathbf{r}_h - \mathbf{r}_{\text{Mn}}) \mathbf{j} \mathbf{S}$ describe the electron-Mn and hole-Mn spin-spin exchange interaction with strengths J_e and J_h respectively, \mathbf{r}_{Mn} is the radius vector of the Mn atom. $\mathcal{H}_{\text{eh}} = -J_{\text{eh}} \delta(\mathbf{r}_e - \mathbf{r}_h) \boldsymbol{\sigma} \mathbf{j}$ is the electron-hole spin interaction Hamiltonian [12]. The Coulomb interaction between electron and hole term is $V_{\text{eh}} = -e^2/\epsilon |\mathbf{r}_e - \mathbf{r}_h|$, where ϵ is the dielectric constant of the system. The last term in Eq. (1) is the Zeeman splitting for the impurity spin.

The electron Hamiltonian in our system is

$$\mathcal{H}_e = \frac{1}{2m_e} \left(\mathbf{p} - \frac{e}{c} \mathbf{A} \right)^2 + V_{\text{conf}}(\rho, z) + \frac{1}{2} g_e \mu_B B \sigma_z, \quad (2)$$

where $\mathbf{A} = \frac{1}{2} B(-y, x, 0)$ is the symmetric gauge vector potential and the last term is the electron Zeeman energy. Without the magnetic field the eigenfunctions of \mathcal{H}_e can be cast in the form

$$\psi_{nl\sigma}^e(\rho, \varphi) = C_{nl} e^{il\varphi} f_{nl}(\rho) \chi_\sigma, \quad (3)$$

where C_{nl} is the normalization constant, $n = 1, 2, \dots$, and $l = 0, \pm 1, \pm 2, \dots$ are the radial and angular quantum numbers respectively, σ is the electron spin and χ_σ is the electron spin wave function. The functions $f_{nl}(\rho)$ are obtained from a suitable linear combination of the Bessel functions

$$f_{nl}(\rho) = J_l(k_{nl}\rho) - \frac{J_l(k_{nl}R_1)}{Y_l(k_{nl}R_1)} Y_l(k_{nl}\rho), \quad (4)$$

where $k_{nl} = \sqrt{2m_e E_{nl}/\hbar^2}$. The corresponding eigenvalues E_{nl} are obtained from the standard boundary conditions of the eigenfunctions.

Taking into account only the Γ_8 states which correspond to the states with the hole spin $j = 3/2$ and include the heavy hole - light hole coupling effects, we construct the single-hole Hamiltonian for the ring as

$$\mathcal{H}_h = \mathcal{H}_L + V_{\text{conf}}(\rho) - 2\kappa \mu_B B j_z. \quad (5)$$

Here \mathcal{H}_L is the Luttinger hamiltonian in axial representation obtained with the four-band $\mathbf{k}\cdot\mathbf{p}$ theory [13, 14]

$$\mathcal{H}_L = \frac{1}{2m_0} \begin{pmatrix} \mathcal{H}_h & R & S & 0 \\ R^* & \mathcal{H}_l & 0 & S \\ S^* & 0 & \mathcal{H}_l & -R \\ 0 & S^* & -R^* & \mathcal{H}_h \end{pmatrix}, \quad (6)$$

where

$$\begin{aligned} \mathcal{H}_h &= (\gamma_1 + \gamma_2)(\Pi_x^2 + \Pi_y^2) + (\gamma_1 - 2\gamma_2)\Pi_z^2, \\ \mathcal{H}_l &= (\gamma_1 - \gamma_2)(\Pi_x^2 + \Pi_y^2) + (\gamma_1 + 2\gamma_2)\Pi_z^2, \end{aligned}$$

$R = 2\sqrt{3}\gamma_3 i\Pi_- \Pi_z$, $S = \sqrt{3}\gamma_1 \Pi_-^2$, $\gamma = \frac{1}{2}(\gamma_2 + \gamma_3)$, and $\mathbf{\Pi} = \mathbf{p} - \frac{e}{c}\mathbf{A}$, $\Pi_{\pm} = \Pi_x \pm i\Pi_y$. $\gamma_1, \gamma_2, \gamma_3$ and κ are the Luttinger parameters and m_0 is the free electron mass.

The Hamiltonian (5) is rotationally invariant. Therefore it will be useful to introduce the total momentum $\mathbf{F} = \mathbf{j} + \mathbf{l}_h$, where \mathbf{j} is the angular momentum of the band edge Bloch function, and \mathbf{l}_h is the envelop angular momentum. Since the projection of the total momentum F_z is a constant of motion, we can find simultaneous eigenstates of (5) and F_z [15].

For a given value of F_z it is logical to seek the eigenfunctions of the Hamiltonian (5) as an expansion [14, 16]

$$\Psi_{F_z}(\rho, \varphi) = \sum_{n, j_z} C_{F_z}(n, j_z) f_{n, F_z - j_z}^h(\rho) e^{i(F_z - j_z)\varphi} \chi_{j_z}, \quad (7)$$

where χ_{j_z} are the hole spin functions and $f_{nl}^h(\rho)$ are the radial wave functions similar to (4) with $k_{nl}^h = \sqrt{2m_0 E_{nl}/\hbar^2(\gamma_1 + \gamma_2)}$. All single hole energy levels and the expansion coefficients are evaluated numerically using the exact diagonalization scheme [16].

In order to evaluate the energy spectrum of the exciton system we need to diagonalize the Hamiltonian (1) without spin interactions in a basis constructed as products of the single-electron and single-hole wave functions. The good quantum number is the projection M_z of the exciton total momentum $\mathbf{M} = \mathbf{F} + \mathbf{l}_e$. For a given value of M_z and the electron spin σ the exciton wave function can be presented as

$$\Psi_{M_z, \sigma} = \sum_{n_e} \sum_{l_e} C(n_e, l_e, F_z) \psi_{n_e l_e \sigma}^e(\rho_e, \varphi_e) \Psi_{F_z}(\rho_h, \varphi_h) \quad (8)$$

The numerical calculations were carried out for a CdTe quantum ring with sizes $R_1 = 100\text{\AA}$, $R_2 = 300\text{\AA}$, $L_z = 30\text{\AA}$ and with the following parameters: $E_g = 1.568\text{ eV}$, $m_e = 0.096m_0$, $g_e = -1.5$, $\gamma_1 = 5.3$, $\gamma_2 = 1.7$, $\gamma_3 = 2$, $\kappa = 0.7$ [17].

To include the spin-spin interactions, we can construct the wave function of the exciton and the magnetic impurity as an expansion of the direct products of the lowest state exciton wave function (8) and eigenfunctions for the magnetic impurity.

$$\Psi = \sum_{\sigma} \sum_{M_z} \sum_{S_z} C(\sigma, M_z, S_z) \Psi_{M_z, \sigma} \times |S_z\rangle. \quad (9)$$

Here $\sigma = \pm 1/2$, $S_z = \pm 1/2, \pm 3/2, \pm 5/2$ and $M_z = \pm 1/2, \pm 3/2, \pm 5/2 \dots$. Using the components of this expansion as the new basis functions we can calculate the corresponding matrix elements for the electron-hole, the electron-impurity and the hole-impurity interactions. Employing the steps used in [7] for the electron-hole spin interaction matrix element, we get

$$M_{eh} = -J_{eh} \delta_{S_z, S'_z} \sum_{j_z, j'_z} A_{eh}(j_z, j'_z) \langle \sigma, j_z | \boldsymbol{\sigma} \mathbf{j} | \sigma', j'_z \rangle, \quad (10)$$

where A_{eh} is obtained by the integration of the electron and hole coordinate wave functions, $\boldsymbol{\sigma}$ is the Pauli spin operator and \mathbf{j} is the hole spin operator [7].

In the case of the electron-impurity interaction we get

$$M_{s-d} = -J_e \sum_{l_e, l'_e} \delta_{M_z - l_e, M'_z - l'_e} A_{s-d}(\mathbf{r}_e = \mathbf{r}_{Mn}, l_e, l'_e) \times \langle \sigma_z, S_z | \boldsymbol{\sigma} \mathbf{S} | \sigma'_z, S'_z \rangle, \quad (11)$$

where A_{s-d} is obtained after the integration of the hole coordinate wave functions and putting $\mathbf{r}_e = \mathbf{r}_{Mn}$ in the electron wave function. Similarly, for the case of hole-impurity interaction we get

$$M_{p-d} = -J_h \delta_{\sigma, \sigma'} \sum_{j_z, j'_z} A_{p-d}(\mathbf{r}_h = \mathbf{r}_{Mn}, j_z, j'_z) \times \langle j_z, S_z | \mathbf{j} \mathbf{S} | j'_z, S'_z \rangle. \quad (12)$$

In order to calculate the spin matrix elements we need to introduce the raising and lowering operators

$$\begin{aligned} \mathbf{S}_+ |S_z\rangle &= \sqrt{S(S+1) - S_z(S_z+1)} |S_z+1\rangle, \\ \mathbf{S}_- |S_z\rangle &= \sqrt{S(S+1) - S_z(S_z-1)} |S_z-1\rangle. \end{aligned} \quad (13)$$

As the spin interactions are short ranged, the most interesting case is when the magnetic impurity is located in the region of average ring radius. In that case we can take $\rho_{Mn} = (R_1 + R_2)/2$ and $\varphi_{Mn} = 0$. The problem was solved numerically using the exact diagonalization scheme and with interaction parameters $J_e = 15\text{ meV nm}^3$, $J_h = -60\text{ meV nm}^3$ [5, 6].

In order to evaluate the optical transition probabilities, let us note that the initial state of the system is that of the magnetic impurity spin with the valence band states fully occupied and the conduction band states being empty. Let us also assume that the impurity states are pure coherent states $|i\rangle = |S_z\rangle$. Recently there were several experimental reports where the quantum dots with a single magnetic impurity in a coherent spin state were prepared even in the absence of a magnetic field [5, 18]. The final states are the eigenstates of the Hamiltonian (1) presented in (9) $|f\rangle = |\Psi\rangle$. In the electric dipole approximation the relative oscillator strengths for all possible optical transitions are proportional to $P(m) \sim |\langle \Psi | m, S_z \rangle|^2$. Here the values of $m = 1, 0, -1$

characterize the polarization of the light as σ^+ , π and σ^- respectively [19]. It should also be mentioned that the impurity spin state remains unchanged during the optical transitions.

In the absence of the magnetic atom in the QR and without the electron-hole spin interaction, the ground state of the exciton will be four-fold degenerate with values of the total momentum ± 1 and ± 2 . The magnetic field lifts that degeneracy due to the Zeeman splitting and as a result two bright ($J_z = \pm 1$) and two dark ($J_z = \pm 2$) exciton states appear. The electron-hole spin exchange interaction in turn gives rise to a further splitting between the bright and dark exciton states and removes the degeneracy between them in zero magnetic field. In Fig. 1 (a) the dependence of few low-lying exciton energy levels on the magnetic field is presented with the electron-hole spin interaction included, for the QR without a magnetic impurity. The corresponding optical transition probabilities for σ^- and σ^+ polarizations are shown in Fig. 1 (b). The sizes of the symbols in Fig. 1 (b) indicate the probability of the optical transition to that state. For smaller values of the magnetic field, two lowest energy levels in Fig. 1 (a) correspond to the dark exciton states and hence the transition probabilities to that states are very weak. The energies of two bright exciton states with the most important components of the basis functions $|\sigma, j_z\rangle = |-1/2, 3/2\rangle$ and $|1/2, -3/2\rangle$ are shifted upwards by the electron-hole spin interaction, but still are clearly visible optically in Fig. 1 (b). In the case of σ^+ polarization we have a strong transition to the state $|-1/2, 3/2\rangle$ (black squares), and for the case of σ^- polarization, the strong transition is for the state $|1/2, -3/2\rangle$ (white squares). It should be also mentioned that with the increase of the magnetic field the transition probabilities remain almost unchanged.

The Mn atom has a spin $S = 5/2$ and there are six possible values of the impurity spin projection S_z . That is why due to the $s, p - d$ spin interaction each exciton energy level presented in Fig. 1 (a) will split into six. In our calculations we consider the energies of first twelve lowest exciton states therefore there are 72 energy levels presented. Due to Zeeman splitting and $s, p - d$ splitting of energy levels there will be many level crossings and anticrossings. The presence of the impurity inside the ring material removes the symmetry of the structure and now we do not have any good quantum numbers to describe the states. All states are mixed superpositions with different values of total momentum of electron, hole and magnetic impurity.

In order to clarify this complicated situation, we have considered here the optical transition spectrum to these 72 states. As we have mentioned above the initial state is a pure coherent state with a fixed value of the impurity spin S_z . The final states are the exciton states with the magnetic impurity. The high probability transitions will be possible only to the bright exciton states, which have

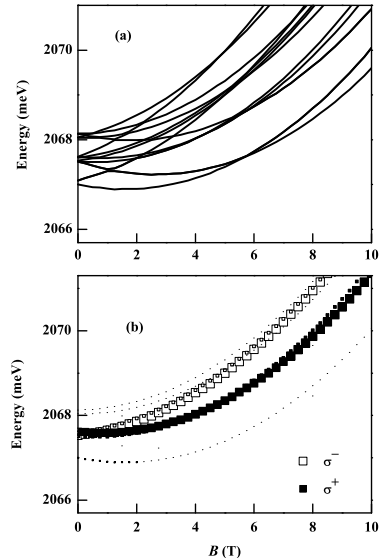


FIG. 1: (a) Magnetic field dependence of the exciton energy levels with electron hole spin-interaction included. (b) Optical transition amplitudes for the σ^+ and σ^- polarizations.

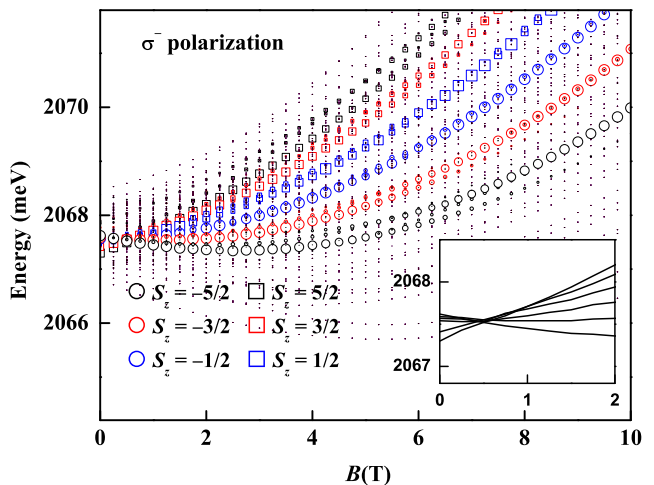


FIG. 2: Magnetic field dependence of the optical transition amplitudes for the case of σ^- polarization and for various values of the initial state impurity spin projection S_z . The crossing point of six bright exciton states is shown as inset.

the most important components with the same value of the impurity spin S_z . The results for the σ^- polarization of the incident light are presented in Fig. 2. Here the shapes and the colors of the points indicate the initial spin of the impurity and the sizes of the points indicates the probability of the transition to that state. For the σ^- polarization of the incident light, the bright exciton states must have the important component with $|\sigma_z, M_z\rangle = |1/2, -3/2\rangle$. For the σ^+ polarization (Fig. 3) the most important component of the bright states must be $|-1/2, 3/2\rangle$ [19]. For example in the case of the

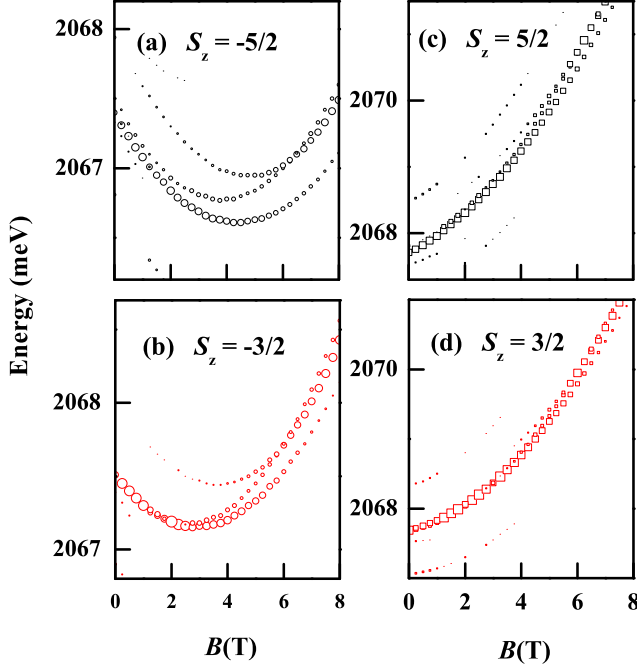


FIG. 3: Magnetic field dependence of the optical transition amplitudes for the case of σ^+ polarization and for various values of the initial state impurity spin projection S_z .

σ^- polarization and for the initial state $S_z = -5/2$ in low magnetic fields we have only one strong transition (Fig. 2 black circles). But near the fields of 5-6 Tesla that line weakens and disappears and a new optical mode appears. Similar behavior can also be seen for other impurity spin states. This effect is the direct signature of the $s, p-d$ spin interaction. Due to spin interactions now the bright exciton state $|1/2, -3/2\rangle$ is coupled with the dark state $|1/2, -1/2\rangle$ and we have two coupled energy levels. For the first level at $B = 0$ the weight of the $|1/2, -3/2, -5/2\rangle$ state is 0.96 and the weight of the $|1/2, -3/2, -5/2\rangle$ state is 0.19. With the increase of the magnetic field the weight of $|1/2, -3/2, -5/2\rangle$ decreases and the weight of $|1/2, -1/2, -5/2\rangle$ state increases. As a result the bright state changes to dark. For the second level we have an opposite picture. In the case of σ^+ polarization (Fig. 3 (a)-(d)) we see similar effects for the case of $S_z = \pm 5/2$ and $\pm 3/2$. Now the bright exciton state $|-1/2, 3/2, S_z\rangle$ is coupled with the dark state $|-1/2, 5/2, S_z\rangle$. For the case of $S_z = \pm 1/2$ the effect is not pronounced because the energies of the mixed bright and dark states are too close to each other.

We should mention here about an interesting effect observed in the case of σ^- polarization. In Fig. 2 there is a crossing point for all energies of the bright exciton states and for $B = 0.5$ Tesla (see inset in Fig. 2). This interesting effect can be explained as follows: In the case of the σ^- polarization the most important component of the bright exciton states is $|1/2, -3/2, S_z\rangle$, where S_z

takes six possible values. For all these states the energy term connected with the $s, p-d$ spin interactions has opposite sign with the Zeeman splitting energy of the magnetic impurity $g_{\text{Mn}}\mu_B B S_z$, where $g_{\text{Mn}} = 2$. For a certain value of the magnetic field B_0 these two terms will cancel each other and we will see a crossing point. In our case $B_0 = 0.5$ Tesla, but in general, the value of B_0 depends on the ring parameters and on the $s, p-d$ interaction constants J_e and J_h . We believe that this effect is experimentally observable. After the detection of the experimental value of the crossing point B_0 one should be able to estimate the real values of the $s, p-d$ interaction constants J_e and J_h in a quantum ring. In the case of the σ^+ polarization the most important component of the bright states is $|-1/2, 3/2, S_z\rangle$. Now the $s, p-d$ interaction term and the Zeeman splitting term for the magnetic impurity always have the same sign and there is no crossing point.

In conclusion, we have studied the effect of spin interactions on the exciton states in a quantum ring with a single magnetic impurity subjected to a perpendicular magnetic field. The optical properties of such a QR have been investigated. It was shown that due to the $s, p-d$ spin exchange interactions between the electron, hole and the magnetic impurity it is possible to change the bright exciton state into a dark state and vice versa with the help of the applied magnetic field. Additionally, a new method is proposed for experimental estimation of $s, p-d$ spin interaction constants.

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- [1] T. Chakraborty, in *Advances in Solid State Physics*, edited by R. Haug (Springer, Berlin, 2003), Vol. 43, pp. 79; T. Chakraborty and P. Pietiläinen, *Solid State Commun.* **87**, 809 (1993); *Phys. Rev. B* **50**, 8460 (1994); *Phys. Rev. B* **52**, 1932 (1995); V. Halonen, P. Pietiläinen, and T. Chakraborty, *EPL* **33**, 377 (1996); K. Niemelä, P. Pietiläinen, and T. Chakraborty, *EPL* **36**, 533 (1996); H.-Y. Chen, P. Pietiläinen, and T. Chakraborty, *Phys. Rev.* **78**, 073407 (2008).
 - [2] T. Chakraborty, *Quantum Dots* (Elsevier, Amsterdam 1999); P.A. Maksym and T. Chakraborty, *Phys. Rev. Lett.* **65**, 108 (1990); T. Chakraborty and P. Pietiläinen, *Phys. Rev. B* **71**, 113305 (2005); *Phys. Rev. Lett.* **95**, 136603 (2005); V.M. Apalkov and T. Chakraborty, *Appl. Phys. Lett.* **78**, 1820 (2001).
 - [3] See, for example, A. Lorke, R.J. Luyken, A.O. Govorov, J.P. Kotthaus, J.M. Garcia, and P.M. Petroff, *Phys. Rev. Lett.* **84**, 2223 (2000); U.F. Keyser, C. Fühner, S. Borck, R.J. Haug, M. Bichler, G. Abstreiter, and W. Wegscheider, *Phys. Rev. Lett.* **90**, 196601 (2003).
 - [4] D. Heitmann, editor, *Quantum Materials* (Springer, Heidelberg, 2010).

- [5] D.E. Reiter, T. Kuhn, V.M. Axt, Phys. Rev. Lett. **102** 177403 (2009); D.E. Reiter, T. Kuhn, V.M. Axt. Phys. Rev. B, **85**, 045308 (2012); D.E. Reiter, V.M. Axt, T. Kuhn. Phys. Rev. B, **87**, 115430 (2013).
- [6] L. Besombes, et al., Phys. Rev. Lett. **93**, 207403 (2004); L. Besombes, et al., Phys. Stat. Sol. (b) **242** 1237 (2005).
- [7] A. Manaselyan, and T. Chakraborty, Nanotechnology **21** 355401 (2010).
- [8] R. Hanson and D. Awschalom, Nature (London) **453**, 1043 (2008).
- [9] L. Besombes et al., Phys. Rev. B **78**, 125324 (2008); A.O. Govorov and A.V. Kalameitsev, Phys. Rev. B **71**, 035338 (2005); Y. Leger et al., Phys. Rev. Lett. **95**, 047403 (2005); Phys. Rev. B **76**, 045331 (2007).
- [10] Ramin M. Abolfath, Pawel Hawrylak, and Igor Zutic, Phys. Rev. Lett. **98**, 207203 (2007).
- [11] T.W. Kim, E.H. Lee, K.H. Lee, J.S. Kim, H.L. Park, Appl. Phys. Lett. **84** (2004) 595;
- [12] A.L. Efros, et al., Phys. Rev. B **54** 4843 (1996).
- [13] J.M. Luttinger, Phys. Rev. **102** 1030 (1956).
- [14] F.B. Pedersen, and Y.C. Chang, Phys. Rev. B **55** 4580 (1997).
- [15] P.C. Sercel, and K.J. Vahala, Phys. Rev. B **42** 3690 (1990).
- [16] A. Manaselyan, and T. Chakraborty, Europhys. Lett. **88** 17003 (2009).
- [17] S. Adachi, *Handbook of physical properties of semiconductors*, Volume 3 (Kluwer, 2004).
- [18] C. Le Gall C, et al., Phys. Rev. Lett. **102** 127402 (2009); L. Besombes, et al., Solid State Commun. **149** 1472 (2009).
- [19] A.K. Bhattacharjee, and J. Perez-Conde, Phys. Rev. B **68** 045303 (2003).